Photoelectron Diffraction of Free CO Molecules

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INTRODUCTION

Core-level photoelectron diffraction techniques have been applied to surface structure studies on an atomic scale for more than 20 years [1]. The basic physical process responsible for the production of a photoelectron diffraction pattern is the elastic scattering of the emitted photoelectron wave. In core level photoemission, a photon illuminates an atom surrounded by other atoms and an electron from an atomic core level is ejected and detected at a distance from the emitter atom. The emitted electron wave may either be detected directly or scattered by neighbouring atoms before detection occures. The interference (diffraction) between the different pathways depends sensitively on the relative atomic positions and the wavelength of the outgoing photoelectron wave. Thus the yield of this process, when measured as a function of photon energy and/or emission angles, contains information about the atomic structure in the vicinity of the emitter.

In the gas phase these effects are only visible if measurements on spatially oriented molecules are performed otherwise the intensity modulations in the electron yield are strongly reduced. So far as we are aware no experiment on this subject has been performed due to difficulties in preparing or determining the spatial orientation of the molecule. In this report we describe an attempt to adapt an experimental setup, already successful in the measurement of the photoelectron angular distribution of spatially oriented molecules [2,3], to photoelectron diffraction studies. The experiment was performed at the beamline 9.3.2 at the Advanced Light Source.

EXPERIMENT

The key element of this experiment is a time-of-flight ion spectrometer with a position sensitive anode optimized for short dead-time [4]. This ion-detector makes it possible to trace the momentum direction of ionic molecular fragments and thus the molecular orientation at the moment of photoionization assuming that the dissociation time is small compared to the timescale of molecular rotation, a condition also known as axial recoil condition. Figure 1 shows a scheme of the experimental setup used for this angular correlation experiment between ionic fragments and photoelectrons.
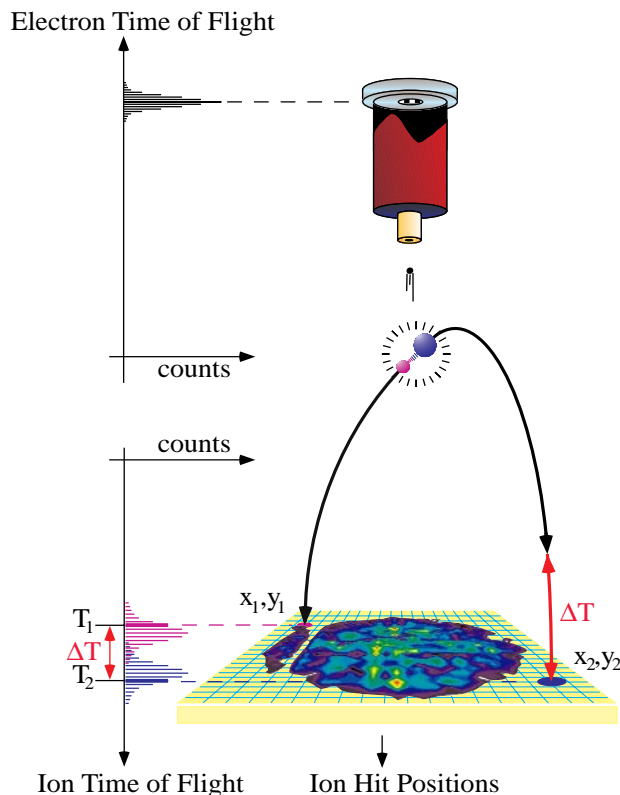

Figure 1. Schematic illustrating the experimental setup with an electron time-of-flight spectrometer (upper part) and an ion time-of-flight spectrometer with a position sensitive anode (lower part).
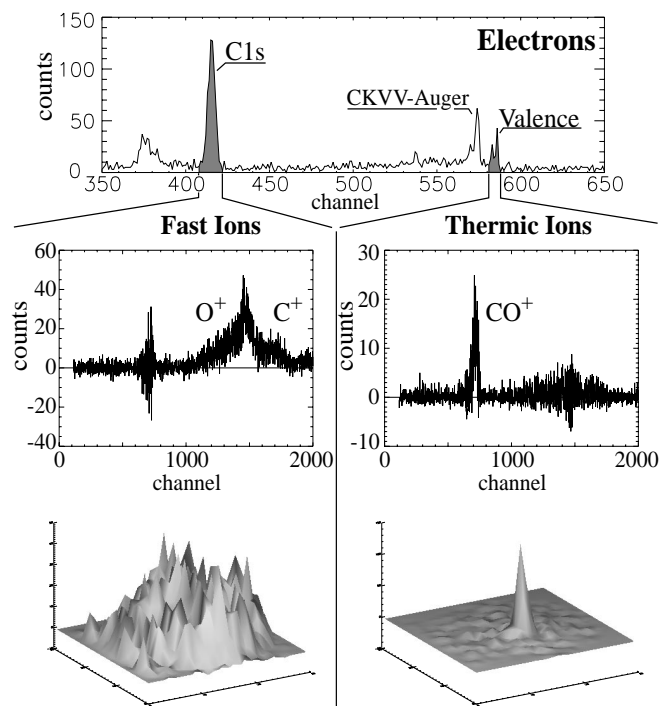
Figure 2. Coincident electron time-of-flight spectrum (upper diagram), ion time-of-flight spectra (middle diagrams) and ion anode hit position distributions (lower diagrams).

The detection of an electron serves as the trigger for an extraction pulse for the ions. Electron time-of-flight, ion time-of-flight, and the ion impact positions on the anode were recorded in coincidence. The resulting experimental coincidence spectra were corrected afterwards for both, uncorrelated and random coincidences by subtraction of normalized spectra obtained by applying pulses from a pulse generator instead of electron signals as a trigger for the ion extraction pulse.

Figure 2 shows some electron and ion time-of-flight spectra of CO at a photon energy of 340.5 eV and the corresponding distributions of hit positions on the ion anode. The upper spectrum is a coincident electron spectrum, while in the middle diagrams ion spectra are shown which correspond to selected kinetic energy ranges of the coincident electrons. The right spectrum shows all ions in coincidence with v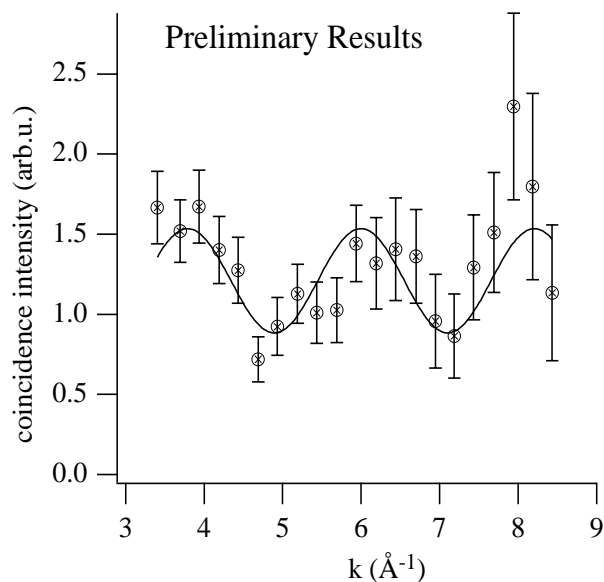alence electrons. These processes lead mainly to stable nonfragmenting CO^+ ions characterized by a sharp peak in the corresponding pattern on the anode. This is clearly visible in a three-dimensional plot of the anode positions below the ion spectrum. Ionic fragmentation following K-shell ionization, into C^+ and O^+ ions in coincidence with the detection of a C(1s) photoelectron (left ion spectrum) yields a two dimensional intensity pattern on the anode (left bottom) which reflects the angular distribution of the fragments. All ion spectra are corrected for uncorrelated and random coincidences.

RESULTS

The ejection angles of the fragment ions were calculated offline using the time-of-flight and the anode hit position information of each electron-ion coincidence event in the experiment. Figure 3 shows the preliminary results of the experiment done in August 1997 during two weeks of double bunch mode of the storage ring. This diagram shows the yield of C(1s) electrons from spatially oriented CO molecules with molecular axes lying within a cone of ±25˚ around the electric vector of light. The electrons were detected along the electric vector of light within

Figure 3. The variation of the C(1s) electron yield detected along the electric vector of light. The emitting CO molecules are orientated in a ±25° cone around the electric vector of light, with the C atom pointing in the direction of the electron detector. The solid line is a simple sin function to guide the eye.

an cone of $\pm 2.6°$. The C^+ ion was pointing towards the electron detector. Figure 3 clearly shows the variation of intensity along the k-value of the outgoing photoelectron resulting from the interference between the direct and scattered photoelectron waves.

In summary, we have shown for the first time quantum interference effects following K-shell photoionization of free CO molecules using a time-of-flight electron spectrometer and a position resolving ion time-of-flight spectrometer. Further analysis of the observed intensity variations is needed in order to reveal the geometrical information which is inherently contained in these data. In future experiments, diffraction patterns of larger molecules may be investigated with respect to electron density distributions which are so far not accessible.

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